THE COMPACTION OF SEMI-PLASTIC MATERIALS

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1. INTRODUCTION

For many years coal research scientists have been trying to find an economic method of briquetting bituminous coal without a binder,[1,2,3,4]. The conventional briquetting processes for coal involve the use of pitch or bitumen as an adhesive; this is undesirable for obvious reasons.

While it has for many years been possible to briquette bituminous coal without a binder, no process has been operated commercially for any length of time, either because the preparation requirements for the coal were too stringent - for example, the coal had to be micronised - or because the compaction pressures demanded were too high for economic operation.

Research at the Coal Research Establishment of the National Coal Board at Cheltenham has sought to remove the need for extremely fine grinding of the particle and the need for the comparatively high pressures of 12 to 15 tons per square inch. A technique has been evolved which works well with British bituminous coals and it has now been applied to many coals from the Commonwealth.

This compaction technique is useful not only with coal: indeed the small financial margins which are available in the coal industries of the world limit this application of the process. After all, where the total process cost cannot exceed 20/- to 30/- per ton, there is no possibility of refined techniques which require machines of some complexity.

It has been shown that the process can be applied to good effect upon catalysts as used in the petroleum and chemical industries, to reactor graphite, iron ore, and many other materials which are loosely termed "semi-plastic".

This paper considers a few aspects of this research and describes some developments which have led to the building of small pilot plant.

2. THE PRINCIPLES OF COMPACTION

In order to obtain a compact of high strength it is necessary to ensure that the compaction technique is effective in bringing about the necessary increase in density and that the compact suffers no damage during extraction from the mould. With coal, at any rate, the strength of a compact for any given size distribution and any given mode of preparation is related to the density of the compact. The density and the strength are determined by the pressure which is applied, but ultimately they approach limiting values which are not exceeded by further increasing the pressure. The limiting density of the compact falls short of the density of the material of the powder by an appreciable margin, say 4 to 20 per cent, depending on the material used (see Fig. 1).

This failure to achieve complete compaction arises in two ways. First, as the briquetting pressure is applied it is opposed by forces set up in the powder and by frictional forces on the walls of the mould which resist the movement of the particles and prevent intimate contact between the surfaces of the particles. Second, when the external pressure is removed the deformed particles recover their shape elastically, in part at least, and the compact expands; the voids within it increase and residual strains remain. A large elastic recovery or a low Young's Modulus is associated with a weak compact.

It has been found that the density of a powder compact can be materially increased for a given applied pressure, if, whilst still under load, it is subjected to shear strain: the strength is increased and the elastic recovery is reduced. The gain in strength with the application of additional shear strain under load may be substantial but the full benefit is obtained only if the shear strain is introduced under maximum load.

The compact made in this way may possess greater density and strength than the limiting values obtained by simple pressing, but whether the compact is made by simple pressing or by introducing additional shear strain under load, the strength and density are still related by the same single curve.

The usefulness of additional shear strain varies with the rheological properties of the material. In the case of a very plastic substance having a negligible elastic recovery, shear strain has little to offer. At the other extreme, anthracite dust or silica sand, both of which are highly elastic and show a very great elastic recovery, say 30 per cent, form no compact at all with or without additional shear strain. The advantage of the process is found to be with materials lying between these extremes, materials which may be termed semi-plastic. Coal is a typical example. The introduction of shear strain improves the briquette by some 5 to 15 per cent as measured by porosity, and the strength is increased by a factor between 3 and 15 (see Figs.1 and 2).

The meaning of shear strain as applied to an elastic body is well understood (ref. 5, and see Fig. 3a) but its precise meaning when applied to a particulate mass is not so obvious. Consider a mass of coal particles in which spherical lead shot are embedded to form a matrix. When pressure is applied to this mass of particles compaction will occur, and at a specific pressure the lead shot will plastically deform to register the deformation of the particles in the neighbourhood of the shot. The spherical shot will have become ellipsoidal and a measurement of their ellipticity will give the angular shear strain which has occurred above the threshold of pressure at which the shot began to deform.

There are several ways in which the ellipticity can be measured and the angular shear strain thus derived. For example, the compact may be made in an aluminium mould which is X-rayed at various pressure levels; the X-ray photographs will show the ellipticity of the shot from which the shear strain can be calculated, or by a somewhat tedious analysis shear strain may be derived from the translation of the shot. Fig. 3b is a series of X-ray photographs showing the deformation of lead shot in such a briquette.

It is possible in the laboratory to make apparatus which will apply a known amount of shear strain more or less uniformly throughout a mass of particles: such an apparatus is a rotary shear box (see Fig. 4) or an annular shear box (see Fig. 5). Experimental work with this type of apparatus permits a fairly accurate examination of the effect of shear strain to be made and is typical of the apparatus used to obtain the compaction curves previously mentioned. There is a theoretical limit to the amount of shear strain that can be introduced;

at high shear strains and with unfavourable stress distributions slip-plane failure results. The onset of slip-plane failure is determined by an equation familiar in soil mechanics:

$$s = \sigma \tan \phi + C$$

where s is the ultimate shearing stress of the briquetted material,
σ is the principal stress normal to the plane of failure,
ϕ is the angle of internal friction,
C is the cohesion of the material.

The locus of the slip plane is formed by a series of points at which the value of s in the above equation is exceeded by the imposed shear stress. An examination of the photograph of the lead shot markers shown in Fig. 3b will show the onset of slip-plane failure at A. Here the slip plane passes through the shot itself, which has been torn apart by the excessive shear stresses present at that point. This photograph also illustrates the very large elastic rebound which causes difficulty in extraction: unless the compaction forces are released evenly the large differential expansion which results will cause obvious or incipient cracking in the briquette. Additional shear strain improves compaction and reduces elastic rebound, but in order to obtain the greatest benefit the additional shear strain must take place at or near the maximum pressure level.

Shear strain applied at pressures below two-thirds the maximum is, in the case of coal, valueless. Fig. 3c shows the effect of shearing at low pressure. For an unsheared specimen the briquetting sequence was compaction under increasing pressure along the curve a to f, followed by elastic recovery and expansion along the curve f, g, as the briquetting pressure was released. For a specimen subjected to a small shear strain early in the briquetting cycle, the sequence was compaction along the curve a to f, as far as b, followed by a reduction in the porosity b to h, whilst shearing takes place at constant pressure. After shearing the briquetting pressure was again increased but the porosity remained substantially constant until the pressure reached the level required to achieve this porosity in the unsheared briquette.

A possible explanation of this is that a specific briquetting pressure is associated with a given area of contact between the coal particles. If this contact area is increased by the application of shear strain, greater pressure can be carried by the particles before they have to bed down further. From this it is seen that to obtain the greatest improvement in briquette quality (as high density implies high strength) additional shear strain must be applied at as high a pressure as possible. If the briquetting pressure is increased after shearing, the briquette may well "forget" that it has been sheared.

The art of making high-density compacts may therefore be summarized as the introduction of the highest possible shear strain at the maximum pressure, the upper limit of shear strain being set by the development of slip-plane failure. This conclusion applies to a wide variety of materials.

3. THE SEVERAL METHODS OF CARRYING OUT THE PROCESS

Two pieces of apparatus of great use in the laboratory have been illustrated in Figs. 4 and 5, but neither of these can be applied to the briquetting of a cheap raw material such as, for example, coal - either because the frictional resistance inherent in the apparatus is too great, or because of mere mechanical complexity, or because the operational sequence is too difficult. Therefore, although these pieces of apparatus produce the most uniform distribution of angular shear strain, and do this at maximum pressure, they are of doubtful value to a commercial process, and alternatives which are theoretically not so desirable have to be used.

The germ of a commercial process is to be seen in a two-stage compaction process in which a mass of particles placed in a mould is first compacted with a plane-ended punch and then with a punch profiled as in Fig. 6. The first compact made in the mould exerts a very considerable lateral thrust on the walls of the mould, and therefore although the plane punch is removed, the particles are still under considerable load when the profiled tool is pressed into the first-formed briquette. The shear strain which then occurs does so under very considerable pressure. This method of manufacture was used to form the lead-shot-marker briquette of Fig. 3b. From this photograph it will be seen that the distribution of shear strain is by no means uniform: the centre core undergoes very high shear strain whereas the corners are friable.

This method of compaction can be developed into a commercial machine in which a composite plunger is used to form a powder into a briquette. Fig. 7a shows a simple form of composite plunger. This has two working parts, an outer annulus and an inner core, which are locked together hydraulically until the average pressure over the cross-section of the briquette has risen to a predetermined level near the maximum (4 to 6 tons/sq.in.). At this point oil is allowed to pass from the locking cavity to a reservoir, the core of the plunger moves forward in relation to the annulus and so causes the required angular shear strain. The setting of the pressure level at which oil is exhausted from the locking cavity is important because shear strain must be made to occur under the highest possible loading. Briquettes made by this duplex plunger possess the qualities of hardness expected from them, but there remains the problem of extracting them from the mould without damage, and this, indeed, poses more difficult problems than those associated with their compaction.

It has been noted already[2] that differential release of load will lead to incipient or obvious cracking of the compact: this, as well as air entrainment, is a frequent cause of "decapping". The only solution is the uniform release of residual strain. An anvil jack is therefore arranged to co-operate with the duplex plunger on its return stroke so that the briquette is subjected to a small longitudinal thrust while it is extracted from the mould. The Poisson ratio effect comes into play and release of strain is very nearly uniform. In the improved plunger shown in Fig. 7b the briquette is made in the extension of the annulus. This arrangement has the advantage that the briquette, when undergoing its initial compression before the application of shear strain, is virtually pressed from both ends so that its density is more uniform and the shear strain, when applied, is more useful. It has the furthe- advantage that extraction is made easier and the anvil jack, essential to the first system, is eliminated. During extraction the centre core of the plunger has to move forward to maintain the longitudinal load upon the briquette at 2 tons $\pm$ 10 per cent while the sides formed by the annulus are withdrawn. This still presents a difficult hydraulic problem.

Each stroke of the duplex ram produces one briquette which may weigh ½ lb. A commercial press has to produce these at many tons per hour. One design of such a press comprises an arrangement of four duplex plungers operating in moulds set in two twelve-station rotary tables. This permits powder to gravity-fill over three stations while it is pressed at another station and ejected at a fifth. The whole assembly is hydraulically operated, tolerances are small and the speed of operation has necessarily to be high. A cycle time of 1 second was the aim set for a large 5 ton/h experimental version illustrated in Fig. 8. It is not yet a reliable machine but it has produced briquettes of excellent quality at 5 ton/sq.in. pressure.

The hydraulic complexity of the turntable presses based on the duplex plunger made it important to develop simpler methods of applying shear strain under load. It was considered wise to sacrifice both the quality of the product and the low power cost to gain simplicity. The expansion press, shown diagrammatically in Fig. 9a is the result of this policy. The briquette emerges as a long rod which has to be broken into suitable lengths; a simple breaker plate can do this.

The pressure/travel curves of the duplex plunger and the expansion press are compared in Fig. 10. The power cost of expansion is approximately twice that of the duplex plunger; this is accounted for by the high frictional resistance of the briquette rod as it moves through the nozzle.

Apart from considerations of mechanical simplicity, power cost and quality of product, the expansion press has one great advantage over the duplex-plunger press - it is insensitive to fill. The duplex-plunger machine, even though it is hydraulically powered, requires that the variation of fill shall not be greater than $\pm$ 3 per cent, whereas the expansion press is equally effective whatever the fill. It is also possible on the expansion press to vacuum-fill; the punch is surrounded by powder which, because the punch is withdrawn at high speed, is forced into the mould under 15 lb/sq.in. pressure and is thereby to some extent compacted. It is surprising that a powder with a free bulk density of 0.4 when fed into a mould in this way has a density within the mould of 0.7.

Because the expansion press is insensitive to fill, and because it is easy to fill the mould, the principle can with comparative ease be applied to a mechanical press (see Fig. 9b) and such a press is probably more suited to the engineering skills available in the briquetting plants of the coal industry than hydraulic presses which need special care in assembly and skill in maintenance. The expansion press is not, however, quite as simple a tool as it appears at first sight. The system is inherently unstable: there is a tendency for the briquetting pressure to drift either up or down so that the machine stalls or briquetting pressure is lost completely. To overcome this, control of the back pressure by a system which is continuous or pulsed is essential. The pressure in the ram applying the lateral force to the four-split flexible nozzle has therefore to be linked via a simple servo control to the pressure developed by the main punch. A typical example of a pulsed control is shown in Fig. 11.

The methods of applying shear strain under load so far described require the use of a reciprocating motion for applying the load, whereas the briquetting of a low-cost raw material is more appropriately carried out in a continuous manner in a double- or ring-roll press. These possibilities have not been overlooked, but it seems unlikely that a pocketed roll can be used to introduce high angular shear strains in this essentially two-stage process. The ring-roll press[3], on the other hand, has some promise. The possibility of causing differential slip between the inner roll and outer ring of such a press has been examined but it is difficult to simulate the feed and discharge arrangements without a fairly large-scale press, and this has not yet been done.

4. PROCESS VARIABLES

Before discussing process variables, the influence of which will vary greatly with the material being compacted, it may be wise to consider the mechanism by which high density in a compact may be achieved. Adhesion may well result from a combination of mechanical interlocking, the development of Van der Waal forces, and hotspot fusion or welding. In any event it is necessary to have substantial plastic deformation of the individual particles, in order either (a) to create new clean surfaces and to bring these surfaces sufficiently close together for molecular forces to develop, or (b) to bring about mechanical interlocking.

Under ordinary conditions, many of the particles from which it is required to form compacts, will fail in compression by brittle fracture. If, however, conditions are such that a hydrostatic state of stress can develop these otherwise brittle particles will deform plastically. It is well known, for example, that a carbon rod under tensile test can be made to elongate and 'neck' in the manner of a mild steel specimen if it is subjected to hydrostatic pressure during test.

The technique which has been described of introducing angular shear strain under load favours the plastic deformation of otherwise brittle material by breaking down internal arches within the particulate mass forming the compact and causing thereby the development of a hydrostatic state of stress. But clearly the process variables should be arranged to give the greatest opportunity for plastic flow to develop: moisture content, temperature, particle size, particle shape, will all play their part in this.

Consider moisture content as a single independent variable; in all probability for a porous material the ease with which a particle plastically deforms will increase with increasing moisture content. But at the same time, excessive moisture on the surface of the particle will form a contaminating film to prevent the surfaces approaching one another, and so developing a molecular bond. For this reason there is likely to be, with many materials, an optimum moisture content. Fortuitously, with most coals, this happens to be at or near inherent moisture content level. Moisture will have a secondary effect as a lubricant.

Rising temperature generally increases the ease with which particles deform and unless this rising temperature is associated with, let us say, oxidation of the surface, which will act as a contaminant, increasing temperature will favour the production of a dense compact. Again there is usually an optimum temperature because some adsorption of gas or vapour often occurs to form a contaminant film.

Particle size is of consequence because small particles are shown by microsquashing techniques to deform more readily than large ones. Surface energy considerations may have a bearing upon this. Generally speaking, finely divided material of laminate form will produce the greatest strength and the highest density.

The photomicrograph (Fig. 12a) of a coal briquette shows the plastic deformation which occurs in the large particles of brittle coal giving rise to a laminate structure. There is evidence that the plastic deformation is such that the surfaces of the particles are brought very close together (see Fig. 12b) and you will note the remarkable difference between the structure of the briquette made with and without additional shear strain, (Figs. 12c and 12d).

Fig. 12e is a photomicrograph of a briquette made at an elevated temperature of 420°C at which the exinite has melted or become very plastic indeed to form a matrix holding the more rigid particles together so forming an interlocked structure of great strength.

Increasing pressure will obviously increase the density of the compact to that point at which internal arching and the frictional resistance on the walls of the mould make further increase profitless. It is obviously of economic importance to use the lowest possible pressure and this variable is not therefore of real importance in this discussion.

Restating the obvious: the greatest density and the greatest strength of compact will be obtained by selecting those conditions of moisture content, temperature, particle size, etc., which favour plastic deformation but which do not give rise to the formation of contaminant film upon the surface of the particles.

5. THE COST OF THE PROCESS

The process was developed for use with bituminous British coal and the financial margin was not greater than 20/- to 30/- per ton. It was therefore of paramount importance to devise machines and a preparation technique which were of the least complication. It was essential to look carefully at existing preparation techniques and to develop high speed special purpose briquetting machines. A sample costing for bituminous coal is given in Table I.

Where the financial margins are not so tight, full advantage can be taken of the substantial gains in strength which are available - in the case of coal it was imperative to use simple robust machines which were unable to introduce the shear strain in a uniform manner - process perfection had to be sacrificed to achieve economic simplicity.

6. CONCLUSIONS

Numerous attempts have been made in the past to briquette bituminous coal without a binder, but no process has been commercially successful. In this case research has devised a process which eliminates the need for fine grinding and high pressure previously essential. Machines have been developed for laboratory use and for commercial use in the coal industry of the United Kingdom.

The compaction technique which is described is applicable not only to coal but to semi-plastic materials generally, and it may be that the effectiveness of the technique can be more completely realised with high cost materials such as catalysts and metal powders, than with coal, where only the most crude application of the process can be contemplated.

7. ACKNOWLEDGEMENTS

The work which is described has been carried out at the Coal Research Establishment, Stoke Orchard, Cheltenham, England, under a programme formulated by the National Coal Board, London, and directed by Dr. D.C. Rhys Jones. It is published by kind permission of the Director-General of Research.

The work reported results from the effort of a small research team which has seen the project through from the laboratory to the pilot plant.

The author is indebted to Dr. B.A. Lilley for information in support of Fig. 3c and to Dr. L. Griffiths for his petrographic studies.

Theories of compaction are many and diverse; the opinions put forward, therefore, in this paper do not necessarily reflect the considered view of the Establishment but weigh heavily upon the author's conscience.

8. REFERENCES

1. Sutcliffe, E.R. U.S. Pat. 1,267,711 (1918). Finely ground coal below 200 B.S.S. mesh subjected to high compressive load.

2. Ten Bosch Octrooien N.V. Brit. Pat. 450,633 (1936). A compression matrix for making briquettes or tablets from fine-grained materials or materials in powder form.
   Ten Bosch Octrooien N.V. Brit. Pat. 440,811 (1936). Method for the manufacture of briquettes from normally non-coherent substances, particularly coal dust, lignite dust and the like.

3. Spooner, E.C.R. Chem. Engng. Min. Rev., 1949, 403, 41. Briquetting by Krupp-Herglotz Ring-Roll Press. Fine grinding of coal and pressures of 12 ton/sq.in. - 17 ton/sq.in.

4. Piersol, R.J. Bull. Ill. geol. Surv., 1948, No. 72; Ill. Geol. Surv. Rep. of Invest., 1936, 41. Pt. I, Smokeless Briquettes; Impacted without Binder from Partially Volatilized Illinois Coals.

5. Timoshenko, S. Theory of Elasticity (New York, McGraw-Hill, 1934).

TABLE I

A Sample of Process Costs - United Kingdom - 1959

These costs refer to a commercial plant forming binderless expansion briquettes 2¾ in. in diameter from high rank coal (volatile matter content 12-20%).

1. Throughput — 125,000 tons/year

2. Capital Costs

Coal Handling	£80,000
Coal drying, grinding, pressing, (including spares)	200,000
Briquette handling	20,000
Instruments	15,000
Site preparation: roads, railway, foundations, buildings, offices, etc.	140,000
10% Contingency	50,000
	£505,000
Interest on capital during construction - 5% for 6 months	£12,000
Working capital, 10% @ 100/- per ton	60,000
Total investment	£577,000
Capital investment per ton/year	£4.6/ton/year

3. Operating Costs (per ton of input)

Wages	2/9d
Power and Heat	4/6d
Repairs and Renewals	1/9d
Overheads and General Expenses	1/6d
Depreciation (plant written off over 10 years)	8/0d
Interest 5%	5/0d
Total cost of carrying out the process	23/6d

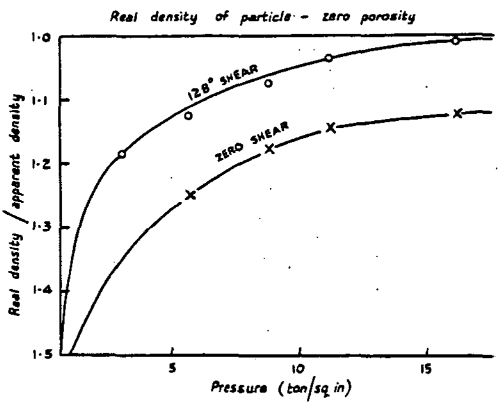


FIG.1. - The relationship between briquetting pressure and density of briquette.

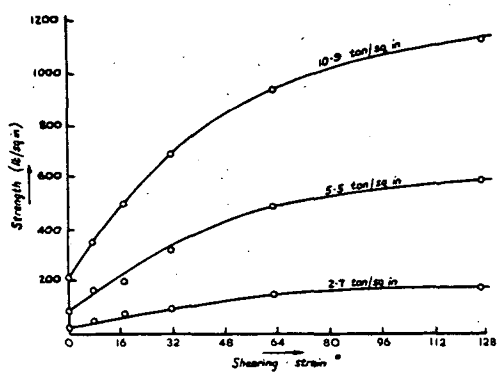


FIG.2. - The variation of briquette strength with applied shear strain.

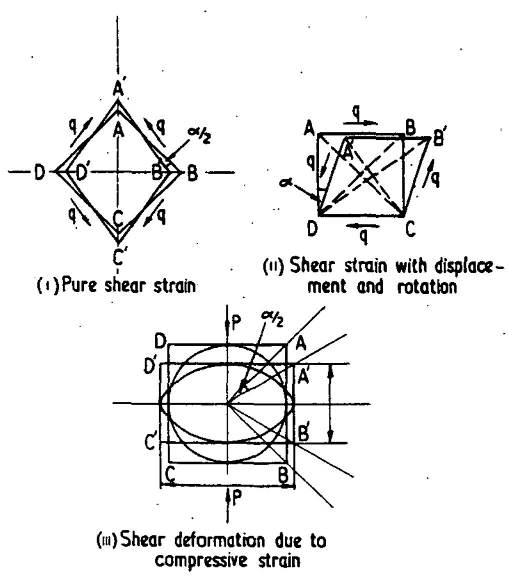


FIG.3(a). - Shear strain - defined as α.

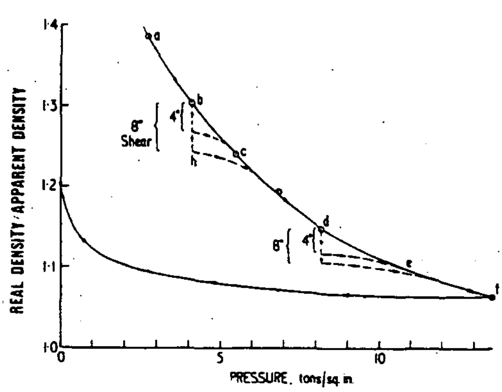
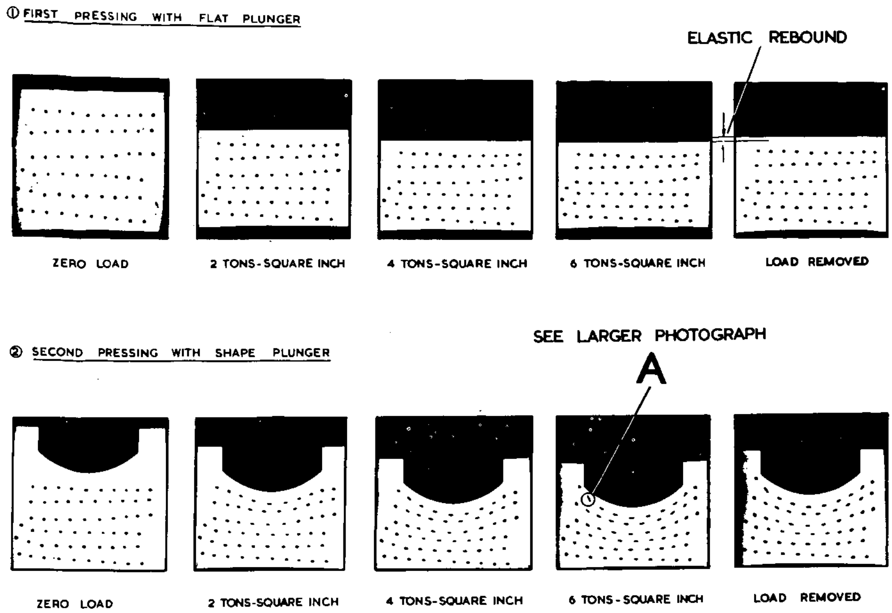


FIG.3(c). - The effect of shearing at low pressure.



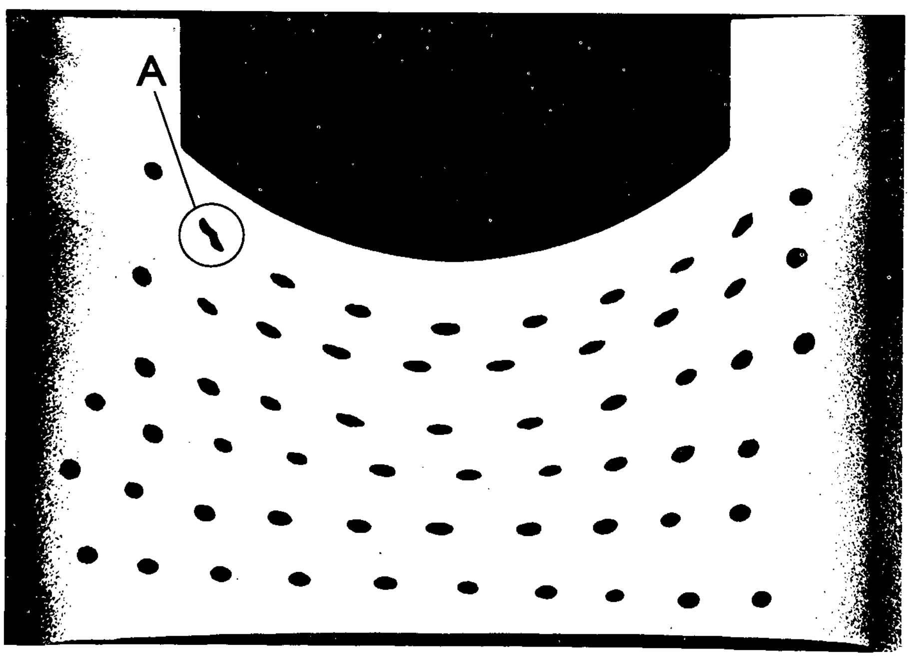


FIG.3(b). - Photograph by X-ray of lead-shot markers in two-plunger briquette.

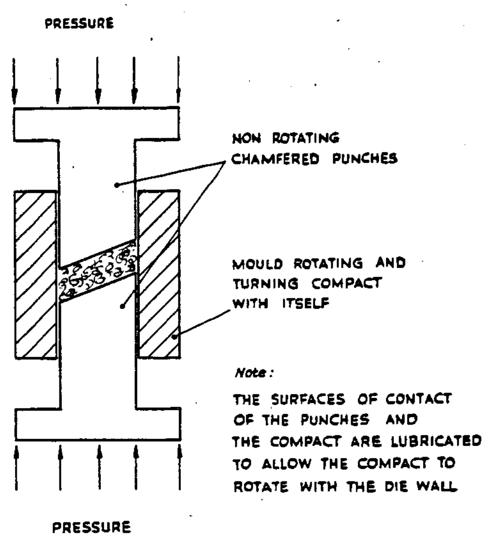


FIG.4. - The rotary shear box.

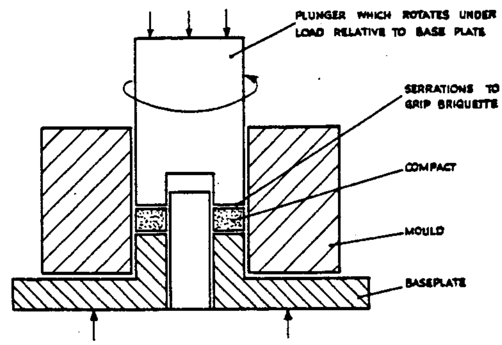


FIG.5. - The annular shear box.

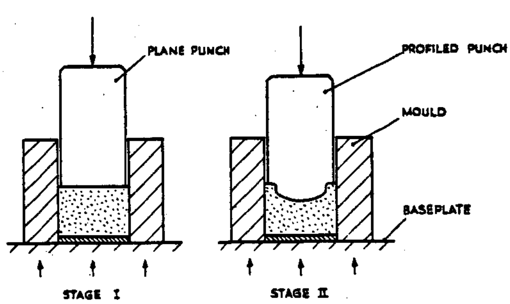


FIG.6. - The double-plunger method.

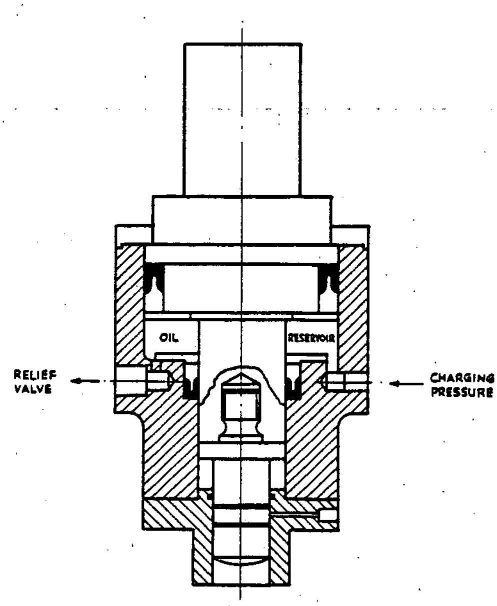


FIG.7(a). - A simple form of composite plunger MK I.

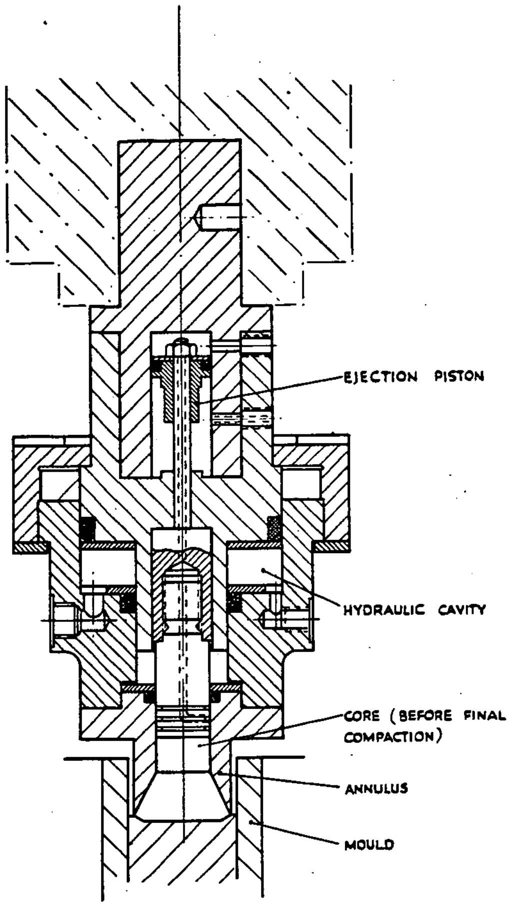


FIG.7(b). - Improved composite or duplex plunger MK II.

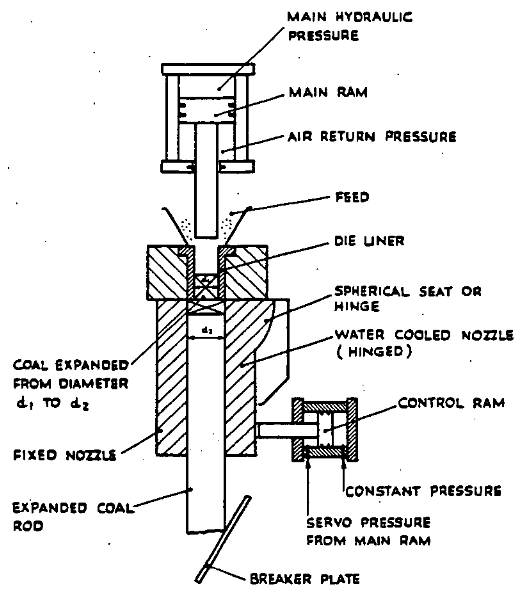


FIG.9(a). - Diagrammatic illustration of expansion principle.

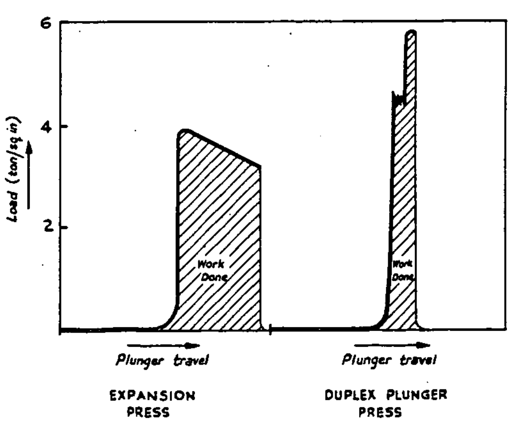


FIG.10. - Pressure-travel curves of duplex plunger and expansion press.

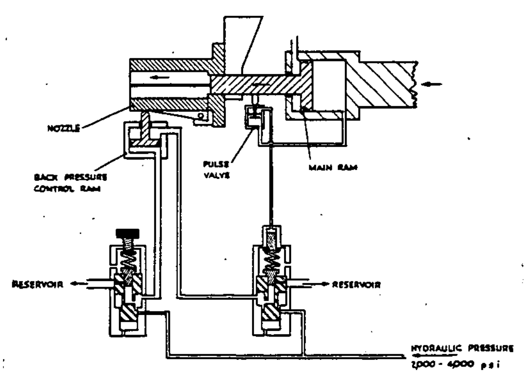


FIG.11. - A back-pressure control system - pulsed control.

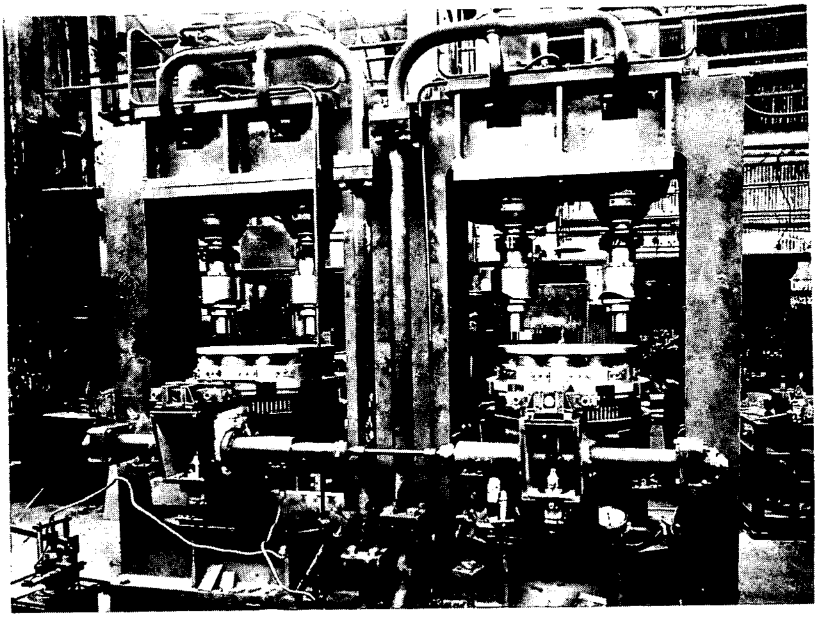


FIG.8 - Hydraulic plunger press.

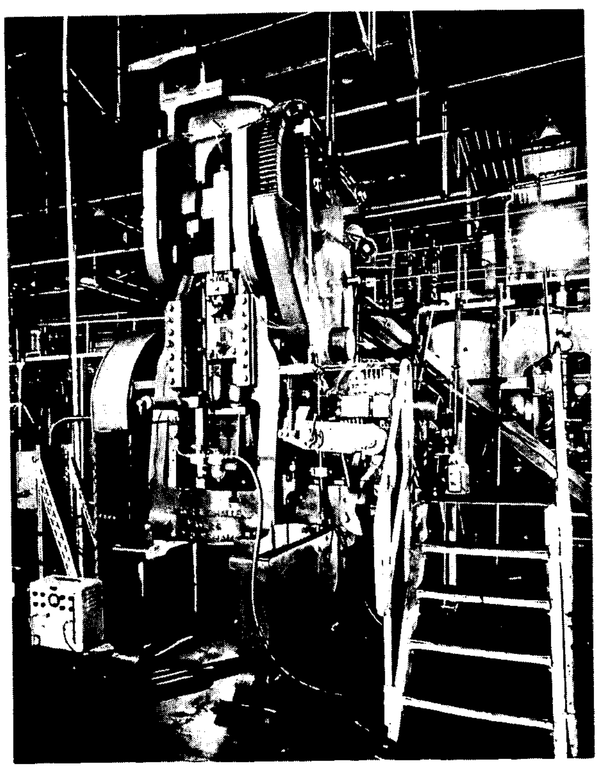


FIG.9(b). - Mechanical expansion press.

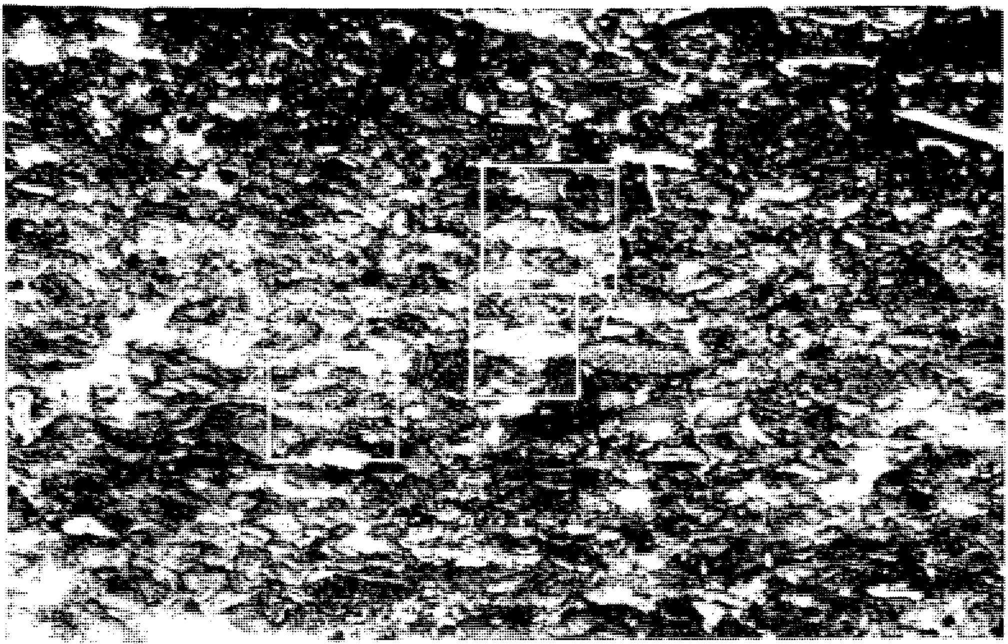


FIG.12(a). - Photomicrograph of Tirpentwys briquette showing deformation of vitrain particles A, B and C and grain. Orientation (dry) magnification 33.

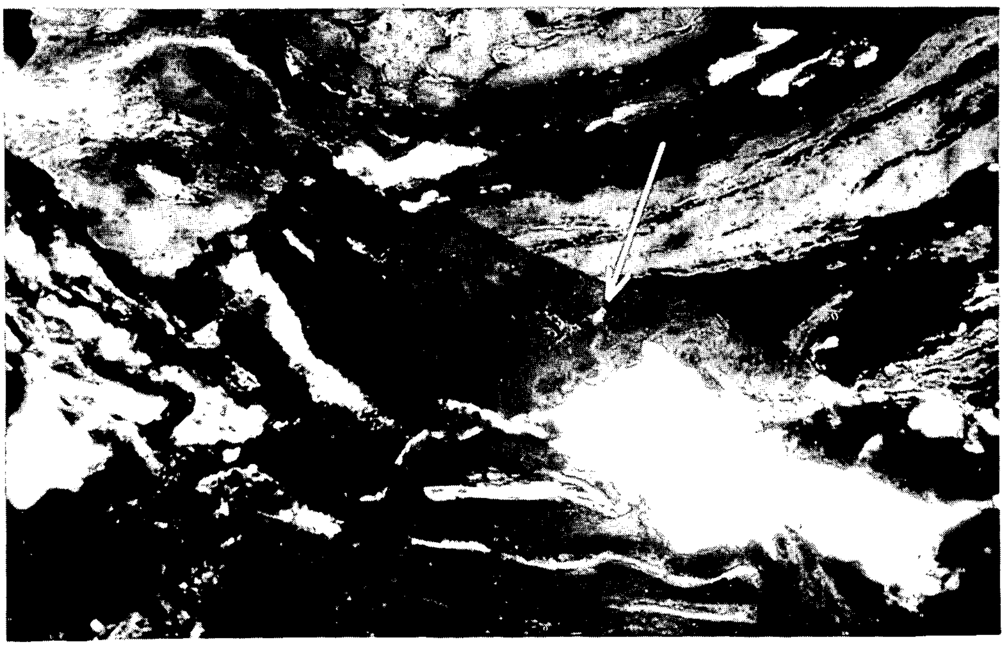


FIG.12(b). - Photomicrograph of Binley briquette made at 110°C showing plastic deformation of vitrain with evidence that the particles are within 0.2 microns (the limit of resolution) (oil, thin section). Magnification 310.

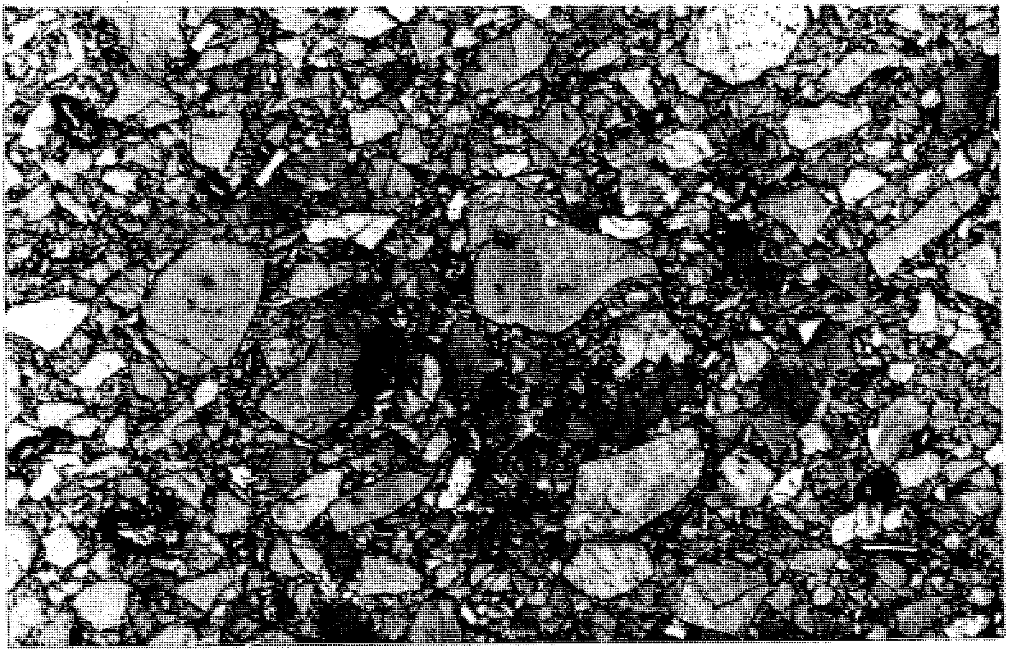


FIG.12(c). - Photomicrograph of coal briquette:
no additional shear strain.

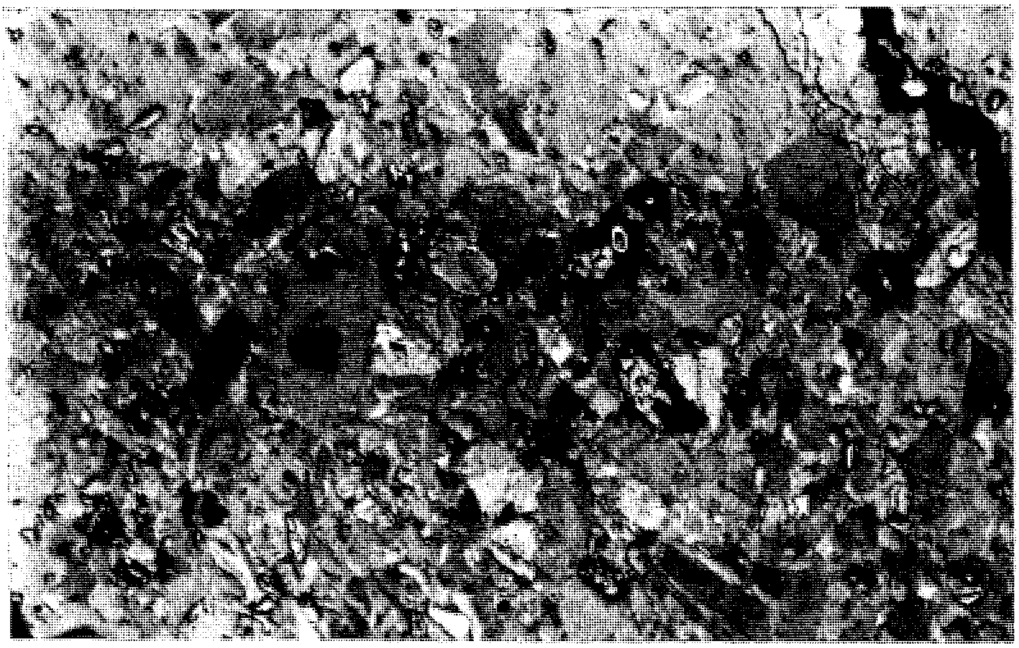


FIG.12(d). - Photomicrograph of coal briquette:
72° of additional shear strain.

FIG.12(e). - Photomicrograph of briquette made at 420°C showing exinite bridging (oil). Magnification 130.